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THE USE OF ELECTRODEPOSITION METHODS TO PREPARE ACTINIDE TARGETS FOR CROSS SECTION MEASUREMENTS AND ACCELERATOR BOMBARDMENTS

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ABSTRACT

The techniques described in this paper were developed in order to produce various actinide targets. A typical target consists of an actinide compound deposited on a metal foil in a layer thin enough to allow various charged particles to pass through the deposit without undue energy loss. The thickness of the deposits ranged from negligible mass up to 1 mg/cm^2. The deposition techniques described may be considered as four separate methods: a) deposition of Pu and Am onto Ni foil from 5 M NH_4Cl solutions, b) deposition of Pu, Cm, Bk, Cf, and Es onto stainless steel foil from isopropyl alcohol-HNO_3 solutions, c) deposition of Cf, Es, and Fm onto Be and Pd foils from dilute HNO_3 solutions, and d) chemical deposition of U onto Al foils. For each method we consider the application for which it was used, the equipment required, the composition of the electrolyte and other experimental conditions, and the advantages and disadvantages of the technique.

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I. INTRODUCTION

The techniques for electrodeposition described in this paper were developed in order to produce actinide targets for fission cross section measurements and charged particle bombardments. A suitable target consists of a thin uniform layer of an actinide compound, chemically pure, deposited on a metal foil. The deposit should adhere well to the foil and be thin enough to allow various charged particles, deuterons, alphas, or fission fragments, to pass through the deposit without undue energy loss. The thickness of the deposits, depending upon the particular experiment, ranged from negligible mass up to 1 mg/cm^2. Since the actinide sample was generally a rare material, e.g. isotopically-enriched ^{242m}Am or the difficult-to-obtain nuclide ^{257}Fm, a very high deposition yield was desired. Electrodeposition was chosen as the technique most likely to produce deposits of acceptable uniformity and adherence with quantitative yield.

In surveying the literature for published methods for electrodeposition of actinides (and lanthanides), many papers were found on the quantitative electrodeposition of tracer-level samples but there were fewer papers on the quantitative deposition of adherent films with the required thickness. The report by Dedov and Kosyakov[1] and the review article by Yaffe[2] provided useful information on electrodeposition techniques for actinides.

Due to the highly positive oxidation potentials of the actinide elements the hydroxide is deposited at the cathode during electrodeposition in aqueous solutions under appropriate conditions. In general it is desirable to electrodeposit from a solution with a reasonably high pH, e.g. in the range 4-5, provided the actinide is sufficiently soluble at these pH values. The high pH minimizes the tendency for the hydroxide deposit to redissolve in the solution, during or just following electrodeposition. This problem is partially alleviated by making the solution basic with conc. NH_4OH just prior to interrupting the current at the end of deposition. We do not thoroughly understand the factors which affect the uniformity and adherence of the hydroxide deposit. The methods given in this report were chosen by trying a number of electrolyte solutions and by varying other parameters, starting with techniques suggested in the literature, until we found a method that produced superior quality deposits with high yield.

The deposition techniques we describe may be considered as four separate approaches: a) deposition from 5 M NH_4Cl solutions, b) deposition from isopropyl alcohol - HNO_3 solutions, c) deposition from dilute HNO_3 solutions, and d) chemical deposition of uranium onto Al foils. For each method we will consider the application for which it was used, the equipment required, the composition of the electrolyte and other experimental conditions, and the advantages and disadvantages of the technique.

II. Electrodeposition from 5 M NH_4Cl solutions.

The neutron-induced fission cross sections of ^{238}Pu, ^{241}Am, and ^{242m}Am were measured in a series of experiments with the Livermore linear electron accelerator acting as a pulsed neutron source. Neutron energies were determined by use of the time-of-flight technique. The target arrangement consisted of a stack of alternating fission samples and spark detectors through which the neutron beam passed with little degradation. Thus, the cross sections for a number of unknown samples and standards were measured simultaneously. The fission fragments escaping from the samples were detected with corona spark detectors which have the property of being insensitive to both α-particle pile-up and γ radiation. A more complete description of the experimental apparatus has been published elsewhere.(3)

In these experiments it was necessary for the plutonium and americium isotopes to be electroplated on both sides of 10-cm diam 0.0076 cm thick Ni plates. A series of tests with different electrolytes and varying experimental conditions were run to optimize both deposition yield and uniformity of the deposit for americium. After trying many systems involving electrolyte solutions of ammonium formate, ammonium oxalate, ammonium carborate, urea, etc., the best results were obtained with samples deposited from 5 M NH_4Cl.

Prior to electroplating, the Ni plate was rinsed with concentrated HNO_3, water, and acetone. After drying, a strippable plastic film(4) was painted on the nickel to define the area on which the americium or plutonium was to be plated. A schematic diagram of the apparatus used for the electroplating is shown in Figure 1. The Ni plate was inserted in a cell which has two circular

(10 cm diam) Pt anodes on either side. The cell is shown in greater detail in Figure 2. The Ni plate was agitated by a vibrator in order to break up gas bubbles which formed in the solution and collected on the plate. This gas, a potentially explosive mixture of hydrogen and oxygen, was removed from the top of the cell by a hood connected to an exhaust line. Cooling of the solution was provided by circulating chilled water on the back side of each Pt anode. Experimental conditions during the plating are summarized in Table I. It is desirable to keep the pH high for the deposition of americium from 5 M NH_4Cl; apparently problems with redissolution of the hydroxide deposit are lessened with resultant improvement in deposition yield and quality of the deposit. For plutonium samples, the acidity of the 5 M NH_4Cl solution was adjusted to pH = 2.4-3.5 prior to the electrodeposition. Attempts to work at higher pH values resulted in the precipitation of plutonium, non-uniform deposition, and low deposition yields.

The electrolyte solution was circulated in and out of the electroplating cell with a pump. The mixing provided by this system produced both an increase in yield and, especially for americium, an opportunity to monitor the concentration of material remaining in the solution. The solution was pumped out of the containment box through polyethylene lines that formed a loop between a NaI detector and the plating cell. The signal from the detector was routed to a single-channel analyzer set to bracket 60 keV and a buffered-memory frequency counter. The count rate is proportional to the concentration of ^{241}Am in solution. A typical series of counts plotted as a function of time is shown in Figure 3. The inclusion of a NaI detector to monitor Am concentration was a vital feature of this system since it permitted a measure of plating efficiency as a function of time.

The deposition yields listed in Table I were determined by assaying the solution before deposition and the solution plus collective rinses after deposition. The gamma counting data were another indication of yield when working with ^{241}Am. The ratio of gamma count rate at the end of the electroplating to that at the beginning should be an indication of the fraction of americium remaining in the solution. In eight separate experiments, the deposition yield deduced from the gamma signal showed an average deviation from the assay value of about 2% in absolute magnitude where the total range of differences was from -3% to +5.5%. Thus, the gamma signal information provides an almost quantitative measure of deposition yield although it may be biased slightly toward high values since there is a tendency for the material being deposited to redissolve once the current has been turned off at the end of the plating. This effect can be seen in the plot of gamma count rate versus time shown in Figure 4 where the current was accidentally interrupted for 20-30 sec. When the plating was nearly complete and the current was about to be turned off, the solution was made basic by adding conc. NH_4OH to avoid redissolution of the deposited sample. After turning off the current, the plate was quickly removed from the solution and the deposit was rinsed with 3 $\underline{M}$ NH_4OH and H_2O. Although a heat lamp was used initially, the highly radioactive deposits seemed to dry quickly without additional heat. The deposits were given no further treatment.

The data listed in Table I are a summary of the preparation of americium and plutonium samples electroplated onto nickel foils in two configurations. In the first, the materials were deposited simultaneously on both sides of the foil in a 10-cm-diam circular deposit; in the second, the materials were deposited on

one side of the foil in a 5.7-cm-diam circular deposit. In the smaller electroplating cell there were some problems initially with non-uniformity in the deposits that was apparently due to using a rotary stirrer. Relatively bare spots appeared in the center of the deposits. Much more uniform deposits were obtained by connecting the stirrer to an electric vibrator where the mixing of the solution is provided by reciprocal motion of the anode. In each of the cells the technique was developed by running a series of ^{241}Am depositions. Data are given in Table I for the best sample yields in these series as well as an average value and range for the deposition yields. It should be emphasized that these techniques gave reproducible results and the lower yields were obtained early in the trial series while certain parameters were being optimized. In each series the average deposition yield was much improved after continuous gamma monitoring of the solution was instituted.

The samples produced for the fission cross section measurements and listed in Table I were not assayed by direct alpha counting. When these samples were made we did not have an appropriate low-geometry counter.[5] As already explained, the deposition yields are known to moderate accuracy based upon assays of the solution. However, absolute assay of the material on the foils was not required since the unknown fission cross sections were determined absolutely by normalizing the data at the energy for thermal neutrons where fission cross sections had already been measured in other experiments.

The uniformity of the samples appeared to be good, based upon visual observations. Cursory checks of activity levels in various locations of each sample indicated moderate but acceptable asymmetry. No quantitative scans of activity versus position on the plate were made. The deposits adhered well to the foils; there was no evidence for loss of material due to flaking.

III. Electrodeposition from isopropyl alcohol - HNO_3 solutions.

A set of heavy element samples were prepared for fission cross section measurements with neutrons in the energy range 20 eV to 10 MeV from an underground nuclear explosion. The techniques used in making these measurements have been reviewed elsewhere.(6) The experiment, designated Physics 8 and executed in August 1969 under the direction of scientists from the Los Alamos Scientific Laboratory, was very successful and resulted in the production of a great quantity of cross section data. At LRL, Livermore, we prepared a series of samples, ^{244}Pu, ^{243}Cm, ^{249}Bk, ^{249}Cf, and ^{253}Es for this experiment. These nuclides are available only in limited quantities and, for the most part, exhibit high levels of α-radioactivity. Because of the high neutron intensity this technique is particularly suited for measuring fission cross sections of microgram samples of intensely radioactive materials. Prior to Physics 8 no detailed measurements of $\sigma_{n,f}$ as a function of neutron energy had been made for any of these species (with the exception of an earlier experiment with ^{243}Cm) although values for thermal neutrons are known for ^{243}Cm, ^{249}Bk, and ^{249}Cf.

In an earlier experiment of this kind (Pommard), we were successful in electrodepositing ^{238}Pu samples from a mixture of isopropyl alcohol and nitric acid. This method produced deposits which appeared uniform and adhered well to the metal backing material. The idea of electrodepositing actinides from mixed solutions involving semi-polar solvents is not new, e.g. Dedov and Kosyakov(1) have shown that use of an alcohol-acetone-HCl mixture produces an adherent plutonium deposit for samples up to a thickness of 300 $\mu g/cm^2$. The isopropyl alcohol-dilute nitric technique was chosen because in tests where plutonium was deposited from dilute HNO_3 the deposits appeared to be less uniform. It was expected that electrodeposition onto stainless steel from 5 M NH_4Cl could result in some chemical attack of the backing material.

The Physics 8 samples were electroplated on 0.00036 cm stainless steel in the form of an ellipse in order to have as much material as possible intercepting a circular beam of neutrons at 45°. Prior to plating, the stainless steel foil was washed with conc. HNO_3, H_2O, and acetone. The design of the electroplating cell is shown in Figure 6. The solution was agitated by connecting the rod supporting the 1.9 cm diam solid Pt anode to a mechanical vibrator. The entire cell was tilted at an angle to allow gas bubbles to escape from beneath the anode. Conditions under which the samples were deposited are summarized in Table II. Prior to preparing these samples, experiments were performed with Cm and Cf tracers and La and Zo carrier material to study variation of yield as a function of certain parameters, i.e. time of deposition, current density, ratio of aqueous solution to alcohol, temperature of solution, etc. All of the samples listed in Table II were electroplated from a mixture of approximately 95% isopropyl alcohol and 5% 0.25 M HNO_3, except for the plutonium solutions which were kept somewhat more acidic to prevent precipitation. A photograph of the ^{244}Pu sample is shown in Figure 7.

The deposition yields in this series were not uniformly high. The yields obtained in producing three ^{244}Pu samples were in the range, 34-45%, considerably lower than the 73% yields obtained while plating a ^{238}Pu sample for the Pennard experiment under essentially the same conditions. A possible explanation of the problem is that some of the ^{244}Pu in the stock solution, 0.5 M HNO_3, had become colloidal and was not electroplated. Since the ^{238}Pu stock solution, 1 M HNO_3, was somewhat more acid, a larger fraction of the plutonium in it may have been in the ionic form. We do feel this technique can be expected to produce high deposition yields for plutonium, although further investigation of the effect of hydrogen ion concentration on deposition yield will be necessary to produce optimum results.

The distribution of material as a function of position in the elliptical deposit was determined by scanning the deposit with a semiconductor alpha detector. The detector, which viewed a 0.13-cm-diam circle on the target foil, scanned across the major and minor axes of the ellipse. Results from scans of ^{249}Cf and ^{249}Bk targets are shown in Figures 8 and 9.[7] The ^{249}Bk deposit was scanned about 20 months after the electrodeposition was performed and, hence, we are counting the ^{249}Cf daughter α-activity from the beta decay of 314-day ^{249}Bk. The α-activity scans indicate a ridge of material on the circumference of the deposit. These peaks near the edges may be resolution limited and may actually be narrower and higher than indicated in the counting. In addition, both the ^{249}Cf and ^{249}Bk deposits display some asymmetry. These α-activity scans are considerably more definitive regarding actual distribution of material than any visual observation. Although these results indicate more inhomogeneity in the deposits than was desirable, the effect of this distribution was of minor importance with regard to any change in solid angle ($< 5\%$) relative to the solid-state fission-fragment detector used in the cross-section experiment.

The α-activity levels of most of the deposits listed in Table II were measured in a low-geometry counter, as shown in Figure 10. The distance between the counting sample and the detector is 76 cm in this system. With a 6.35-mm-diam collimator in place, the counter has a geometry of 4.41×10^{-6} for a point source; the geometry for the actual elliptical sources is calculated to be larger than that for a point source by 1 part in 18,000. The geometry of the counter, as calculated from the dimensions of the chamber and collimator, was checked with an independently calibrated ^{241}Am source;[8] the difference in the two values was 0.7%. An independent assay of the ^{249}Cf sample at the Los Alamos Scientific Laboratory was in agreement

with our value to 1%.[9] With a 3.28-mm-diam collimator, the geometry is low enough (1.1×10^{-6}) to allow counting of the ^{253}Es source, 2.3×10^{11} α dpm. The chamber is large enough to permit counting of samples up to 28 cm in diameter.

The samples listed in Table II all demonstrated good adherence to the stainless steel backing. Of course, care was taken not to rub the deposit directly. There is good agreement between assays of the ^{249}Bk, ^{249}Cf and ^{253}Es samples taken 30 days apart, before and after the experiment (see items in Table II with references to footnotes d, e, and i). Thus, there was no loss of material during the shipping of samples from LRL to the Nevada Test Site and Los Alamos and return or during the handling before and after the experiment. Reassay of the ^{243}Cm sample following the experiment indicated a 4% loss, although the circumstances of the loss are uncertain since there was no evidence of contamination in the shipping container or during the handling of the foil.

Direct alpha counting of each sample is clearly a desirable method of assay. In some instances the yield of electrodeposited material was determined indirectly by assaying the contents of the plating solution before and after deposition. A comparison of results from both methods is shown in Table II. The best agreement was obtained for the ^{249}Cf (II) sample; poorer agreement was found for the ^{249}Bk sample. The indirect determination proved to be least accurate in the preparation of the plutonium samples, especially ^{244}Pu. The deposition yield indicated by assay of the solution after plating was much higher than actually obtained. This result is consistent with the suggestion that part of our plutonium was in colloidal form and was unavailable for electrodeposition or post-deposition assay. Since we were unable to obtain a satisfactory material balance, we can only suggest a possible mechanism for loss.

Another method of assay is to carefully dissolve all of the deposited material following completion of the experiment and measure the concentration of the resultant solution. In the case of the ^{249}Cf (I) sample, the difference between direct alpha counting and assay of the dissolved sample was 1.3%.

IV. Electrodeposition from dilute nitric acid solutions.

The electrodeposition of actinide elements from dilute nitric acid solutions (pH $\approx$ 3) has been reported to be a useful method for producing both tracer level and thicker deposits of americium and curium.[1] We have used this method to produce Cf, Es, and Fm targets for charged-particle bombardments. In these experiements it is desireable to produce as thin and uniform a deposit as possible because the recoil products from (α,xn) and (d,p) reactions must escape from the target. One of the experiments involved collecting ^{258}Fm recoil atoms during the bombardment of a ^{257}Fm target.[10] Since the ^{258}Fm was produced by a (d,p) reaction with incident 12 MeV deuterons, the recoil atoms had very little momentum. Thus, a particularly thin target was necessary.

The preparation of a 0.001 M HNO_3 electrolyte solution is simple enough, although it is not always easy to completely dissolve the target material in a small volume of weak acid. Inability to obtain complete dissolution can be an important limiting factor in the overall yield in producing the target. At this pH there may also be some losses of material due to adsorption onto glass surfaces. However, use of the experimental apparatus[11] shown in Figure 11 with this method for electrodeposition has proven to have several advantages: a) it is relatively easy to handle small volumes of solution, b) it is useful in producing small diameter deposits, c) the electrolyte solution contains no salts and, hence, the deposition can be repeated without further chemical purification.

The actinide to be electrodeposited has often just been purified; a late step in the chemical procedure is usually elution from a cation exchange column. The solution is then evaporated dry, 1 drop of $HClO_4$ is added and fumed to dryness, 2 drops of 1 M HNO_3 are added, and the sample is again dried. This treatment insures

destruction of any residual cation-exchange resin. After dissolving the material in 0.001 M HNO_3 the sample is ready for electrodeposition. The volume of solution used in the apparatus is small, ranging from 0.05 ml to 0.2 ml depending upon the diameter of the glass tube.

The experimental arrangement for use of these targets at the accelerator required the backing material to be relatively thin (the incident beam passes through the backing material before entering the target) and to be able to withstand high temperatures; thin Be foils (0.001-0.002 cm thick) were found to be best for this purpose. Several deposits were placed directly onto Be after degreasing the foils with CCl_4; however, some Be foils did not remain passive during the electroplating and were attacked by dilute HNO_3. An unreactive plating surface was produced by vapor-depositing a thin coating of Pd (100-200 $\mu g/cm^2$) onto the Be.

The plating cell is a short glass tube which rests on top of the metal target foil (see Figure 11). The bottom surface of the glass tube has been ground to provide a smooth surface in contact with the metal foil. This tube is connected to a four-way glass joint by a piece of tygon tubing. The upper vertical section of the glass joint has a Pt wire sealed through the top. The Pt wire serves as an anode and extends down into the solution to within 1-2 mm above the plating foil. One horizontal arm is connected to a syringe while the other has a U-tube filled with water. The pressure above the electrolyte solution can be controlled by movement of the syringe. During the electroplating it is necessary to slowly withdraw the plunger of the syringe; otherwise, gas evolved from the decomposition of H_2O would force the solution out of the cell. This arrangement for controlling the liquid has proven to be quite convenient, e.g. it is a simple matter to remove the plate after deposition is complete. After reducing the current to cut down on

gas evolution, the stand and plate are lowered a few mm. While maintaining slightly reduced pressure above the liquid, the plate may be carefully moved so that the deposit is no longer under the cell nor is there any solution left on the deposit. The current is then turned off and the electrolyte solution is removed with a transfer pipette. The new position on the metal plate to which the cell was moved is rinsed with a small amount of acid and H_2O.

The targets produced by use of this technique are listed in Table III. The ^{253}Es target was used in 11 separate α-particle bombardments in which it received a total of 1500 μamp-hr irradiation. During the first 10 of these bombardments recoil atoms of Md and Fm were collected from the target with efficiencies of 40-50%. Since the collection efficiency did not seem to decrease appreciably with time, the condition of the target was apparently stable. In the work with the ^{257}Fm targets, the recoil collection efficiencies were observed to decrease from an initial 40% to about 6% after many bombardments. This decrease was attributed to sputtering of metal and deposition of decomposition products from organic vapors onto the target rather than a change in the Fm deposit. Thus, the targets prepared by the method described in this section exhibit sufficient uniformity and adherence to be used satisfactorily in accelerator bombardments where one wants to collect recoil atoms efficiently.

V. Deposition of uranium by chemical replacement.

It has been reported by Skorka[12] that uranium is readily deposited on aluminum by chemically replacing the aluminum ions with uranium ions. The reaction proceeds according to the equation:

$$3\ UO_2^{++} + 2\ Al + 2\ H_2O \longrightarrow 2\ Al^{+++} + U_3O_8 + 2\ H_2$$

The electromotive force used to deposit the uranium is not applied externally; instead, the free energy of the above reaction favors deposition of U_3O_8 on the aluminum surface. The uranium is deposited in a dull black, adherent layer. Skorka states that the temperature of the solution must be above 75°C and that the aluminum should be sanded beforehand with emery paper to remove the oxide layer, but not treated with any acids or bases. He recommends deposition from uranyl nitrate solutions with concentrations in the range 5-10 g/ℓ.

We have developed our own version of this technique,[13] with modifications of the method described in Skorka's brief paper.[12] The technique has been employed, in lieu of various electrodeposition methods, to produce deposits of uranium provided the experimental requirements permit the use of Al backing material and there is sufficient uranium that deposition yields of ~ 5% are acceptable. When an experiment requires plating uranium onto an irregularly-shaped object, we have found this technique exceptionally useful whereas uniform electrodeposition may be more difficult. For example, neutron detectors of various designs often employ deposits of uranium in many configurations, plated on the inside, outside, or both surfaces of cylinders.

Briefly, our procedure is the following: The uranium is dissolved in an HCl solution with the pH adjusted to 2.7-3.2 with dilute NaOH. (Skorka suggested deposition from uranyl nitrate solution with no adjustment of pH.) The NaOH should be

added dropwise very slowly with stirring to avoid causing the uranium to precipitate. The solution is heated to 90°C. The aluminum backing material is cleansed of any grease with CCl_4 and then etched with 20% NaOH for 20 seconds. The aluminum is then rinsed with H_2O and etched with 3-6 M HCl for 20 seconds. Immediately after removing the aluminum from the HCl solution and rinsing with H_2O, the Al backing material is introduced into the hot uranium solution. There should be no delay at this point or the Al surface will begin to oxidize with consequent loss of yield and uniformity in the deposit. The deposition yield will be determined by the length of time the aluminum is left in the uranium solution. After 1 min about 1.5% is deposited; after 3 min about 5% is deposited. Usually the maximum deposition yield that can be obtained is 7-10%, regardless of the length of time the Al remains in the solution. There is also a limiting thickness of about 3 mg/cm^2 regardless of the initial concentration of uranium in the solution; adherence becomes a problem for deposits thicker than 1.5-2 mg/cm^2.

It is best to use Al backing material that is at least 0.025-cm thick so that it may be thoroughly etched prior to deposition in order to give the most uniform and adherent deposits. For certain experiments where a thinner backing is desired, we have successfully produced 20-cm-diam circular deposits of 750 $\mu g/cm^2$, high purity ^{235}U, on 0.0025-cm-thick Al foil. In order to work with thin foils, the usual technique is modified by using more dilute solutions and shorter times during the predeposition etching. These thin foils should be free of small holes since any holes are enlarged during etching.

Indirect determination of the uranium deposited can be made in the manner common to all deposition techniques; i.e. measuring the uranium concentration and total volume of the solution before and after deposition.

REFERENCES

1. V. B. Dedov and V. N. Kosyakov, Proceedings of the Int'l. Conf. on Peaceful Uses of Atomic Energy 7, 369 (1956).

2. L. Yaffe, Annual Review of Nuclear Science 12, 153 (1962).

3. C. D. Bowman, G. F. Auchampaugh, S. C. Fultz, and R. W. Hoff, Physical Review 166, 1219 (1968).

4. "Marliner" strippable film (NEC Equipment Corp., Newton Highlands, Mass.).

5. A suitable low-geometry counter was later designed and built. It is described in Section III.

6. B. C. Diven, Annual Review of Nuclear Science 20, 79 (1970).

7. We are indebted to Dr. M. G. Silbert for making these measurements and allowing us to present the data shown in Figures 8 and 9.

8. The ^{241}Am reference source was calibrated at LRL, Berkeley, in a low-geometry counter of precisely-known geometry. We are indebted to Mr. H. P. Robinson for this measurement.

9. We are indebted to Dr. M. G. Silbert for an independent assay of the ^{249}Cf sample.

10. E. K. Hulet, J. F. Wild, R. W. Lougheed, J. E. Evans, B. J. Qualheim, M. Nurmia, and A. Ghiorso, Physical Review Letters 26, 523 (1971).

11. We are indebted to Dr. F. Asaro for providing a prototype of the electrodeposition cell and discussions of his experience in its use. Mr. R. M. Latimer is responsible for development of the technique at the Berkeley laboratory.

12. S. Skorka, Naturwissenschaften, Vol. 40, No. 23, 605 (1953).

13. We are indebted to Mr. F. B. Stephens for his ideas and assistance in developing this chemical plating method.

TABLE I Electrodeposition of samples from 2% NH_4Cl solutions

Material deposited	Amount and composition of material on plate	Thickness of deposit ($\mu g/cm^2$)	Electrolyte composition	Current density (amp/cm^2)	Voltage (volts)	Time of deposition (min)	Deposition yield	Remarks
^{242m}Am,^{241}Am	53.7 mg (19.8% ^{242m}Am, 79.5% ^{241}Am, 0.7% ^{243}Am)	350	2% NH_4Cl, pH = 5.1[a]	0.37	3.8-3.9	40	96%	Footnote b.
^{241}Am	75 mg (100% ^{241}Am)	460	2% NH_4Cl, pH = 5.1[a]	0.37	3.6-3.7	30	6 separate runs: best yield 97% average yield 92% range 82-97%	Footnote b.
^{239}Pu	156 mg (99.3% ^{239}Pu)	960	2% NH_4Cl, pH = 2.4[c]	0.39	3.5	33	94%	Footnote b.
^{238}Pu	26 mg (99.88% ^{238}Pu)	160	2% NH_4Cl, pH = 3.5[c]	0.37	4.0	40	5 separate runs: best yield 87% average yield 64% range 36-87%	Footnote d.
^{242m}Am,^{241}Am	23 mg (19.8% ^{242m}Am, 79.5% ^{241}Am, 0.7% ^{243}Am)	880	2% NH_4Cl, pH = 5.1[a]	0.38	5.0	26	91%	Footnote e.
^{242m}Am,^{241}Am	24 mg (19.8% ^{242m}Am, 79.5% ^{241}Am, 0.7% ^{243}Am)	920	2% NH_4Cl, pH = 5.1[a]	0.38	4.8	27	94%	Footnote e.
^{241}Am	25 mg (100% ^{241}Am)	960	2% NH_4Cl, pH = 5.1[a]	0.38	4.9	26	10 separate runs: best yield 98% average yield 76% range 25-98%	Footnote e

Footnotes for TABLE I

a - The pH of the solution was adjusted by adding methyl red indicator, making the solution just basic with 3 M NH_4OH, and then acidifying with 3 M HCl until the indicator turned pink.

b - Large cell: Electrodeposition on both sides of foil in 10 cm diam circular deposit; total area 162 cm^2; foil 0.0076 cm thick Ni; anode-cathode distance 0.47 cm; solution volume 260 ml; solution temperature 28°C; see Fig. 2.

c - The pH of the solution was determined by use of a pH meter.

d - Conditions are the same as in footnote b except the foil is 0.013 cm thick Ni.

e - Small cell: Electrodeposition on one side of foil in 5.7 cm diam circular deposit; total area 26 cm^2; foil 0.025 cm thick Ni; anode-cathode distance 1 cm; solution volume 55 ml; solution temperature 28°C; see Fig. 5.

TABLE II - Electrodeposition of samples from isopropyl alcohol - HNO_3 mixtures.

Material deposited	Isotopic composition	Assay of material on plate (μg)	Activity level	Thickness of deposit (μg/cm²)	Electrolyte composition	Solution temperature (°C)	Current density (amp/cm²)	Voltage (volts)	Time of deposition (min)	Deposition yield	Remarks
^{244}Pu	99.08% 244	588 [a]	4.6×10^5 α dpm	518	0.6 ml 0.5% HNO_3 + 10 ml isopropyl alc.	57	0.053	300	30	3 separate runs: best yield 49% average yield 38% range 34-49%	Other conditions for all samples listed in this table are given in footnote b.
^{243}Am	89.0% 243, 10.9% 244	212.5 [c,d] 204.2 [c,e]	2.4×10^{10} α dpm	184	0.5 ml 0.2% HNO_3 + 9.5 ml isopropyl alc.	39	0.044	---	30	70 - 80% [f]	
^{249}Bk	97.5% ^{249}Bk, 2.5% ^{249}Cf [g]	(98.7) [d,h] 98.5 [i]	3.7×10^{11} β dpm 8.5×10^7 α dpm	87	same as above	39	0.044	160-230	38	97%	
^{249}Cf (I)	100% 249	95.0 [c] 96.8 [j]	8.6×10^8 α dpm	84	same as above	39	0.044	180-205	89	84%	
^{249}Cf (II)	100% 249	(99.8) [d,h] 100.7 [c,d] 100.9 [c,e]	9.1×10^8 α dpm	89	same as above	39	0.044	185-210	28	90%	
^{253}Es	63% ^{253}Es, 37% ^{249}Bk-^{249}Cf [k]	3.26 [c,d] 3.38 [c,e]	1.8×10^{12} α dpm	2.9	same as above	39	0.044	125	35	~0%	
^{238}Pu	99.99% 238	1,120 [c]	4.3×10^7 α dpm	987	0.5 ml 1% HNO_3 + 10 ml isopropyl alc.	57	0.082	300	15	~3%	Footnote i

FOOTNOTES FOR TABLE II

a - Direct assay by α-counting in 2π geometry.

b - Electrodeposition on one side of foil in elliptical shape, 1.4 cm by 1.0 cm; total area 1.13 cm^2; foil 0.00036 cm thick stainless steel; anode-cathode distance 2 cm.

c - Direct assay by α counting at low-geometry (geometry factor = 4.41×10^{-6}).

d - Amount of element before experiment, corrected for decay to 8/27/69.

e - Amount of element after experiment, corrected for decay to 8/27/69.

f - The exact yield is uncertain due to problems in assay of the initial solution.

g - Composition as of 8/27/69, 12 days after Bk purification.

h - Indirect determination of material on plate by assaying the content of the plating solution before and after electrodeposition; this method is considered less accurate than direct assay.

i - Direct assay in low geometry counter; observed growth of ^{249}Cf α activity following Bk purification; data taken before and after experiment indicate loss ≤ 0.5% during handling and shipment of target.

j - Determination made by complete dissolution of the sample followed by radiometric assay of the solution and α-counting in a 2π counter.

k - Composition as of 8/27/69, 14 days after Es purification.

ℓ - Conditions for electroplating this sample were the same as for the ^{244}Pu sample above; this sample was used for fission cross section measurement in the Pommard experiment conducted in March 1968.

TABLE III. Electrodeposition from dilute HNO_3 solutions.

Material deposited	Amount of target material (μg)	Thickness of deposit (μg/cm²)	Current density (amp/cm²)	Voltage (volts)	Time of deposition (min)	Deposition yield[a]	Remarks
^{243}Am tracer + La carrier	10[b]	56	0.20	100	20	8 separate runs best yield 87% average yield 76% range 64-87%	Footnote c
^{252}Cf	2.5	14	0.11-0.22	60-90	27	56%	Footnote c
^{253}Es	2.7	15	0.11-0.17	125	6	70%	Footnote c
^{257}Fm I	2×10^{-6}	5.3×10^{-5}	0.26-0.39	55	8	47%	Footnote d
II	Same as above		0.26-0.52	125	8	75%	Footnote e
III	Same as above		0.39	75	7	73%	Footnote e
IV	Same as above		0.39	80	7	89%	Footnote e

a - The yield was measured indirectly by radiometric assay of plating solution before and after electrodeposition.

b - Amount of La carrier.

c - Electrolyte 0.001 M HNO_3; electrodeposition on one side of foil in 0.48 cm diam. circular deposit; total area 0.18 cm²; foil 0.0025 cm thick Be; anode-cathode distance 1-5 mm; solution volume 0.2 ml.

d - Electrolyte 0.001 M HNO_3; electrodeposition on one side of foil in 0.22 cm diam. circular deposit; total area 0.038 cm²; foil 0.0013 cm thick Be; anode-cathode distance 1-5 mm; solution volume 0.05-0.1 ml.

e - Conditions are the same as in footnote d except that the Be foil had a 100-200 μg/cm² coating of Pd where the target was electrodeposited.

FIGURE CAPTIONS

1. Schematic of equipment used to electrodeposit 10 cm diam samples of Pu and Am from 5 M NH_4Cl solutions onto both sides of Ni foils.

2. Electroplating cell used to deposit 10 cm diam samples of Pu and Am from 5 M NH_4Cl solutions to both sides of Ni foils.

3. Data from NaI detector used to monitor concentration of ^{241}Am in solution - plot of gamma count rate versus time.

4. Data from NaI detector used to monitor concentration of ^{241}Am in solution - plot of gamma count rate versus time. Current interrupted during run.

5. Electroplating cell used to deposit 5.7 cm diam samples of Pu and Am from 5 M NH_4Cl solutions onto one side of a Ni foil.

6. Electroplating cell used to deposit samples of Pu, Cm, Bk, Cf, and Es from isopropyl alcohol - HNO_3 solutions onto stainless steel foils in an elliptical shape.

7. Sample of ^{244}Pu (588 μg) for Physics 8 experiment.

8. Scan of alpha activity across major and minor axes of ^{249}Cf deposit for Physics 8 experiment.

9. Scan of alpha activity across major and minor axes of ^{249}Bk deposit for Physics 8 experiment.

10. Low-geometry alpha counter.

11. Electroplating equipment used to deposit 2-5 mm diam samples of Cf, Es, and Fm from 0.001 M HNO_3 solutions onto Be foil.

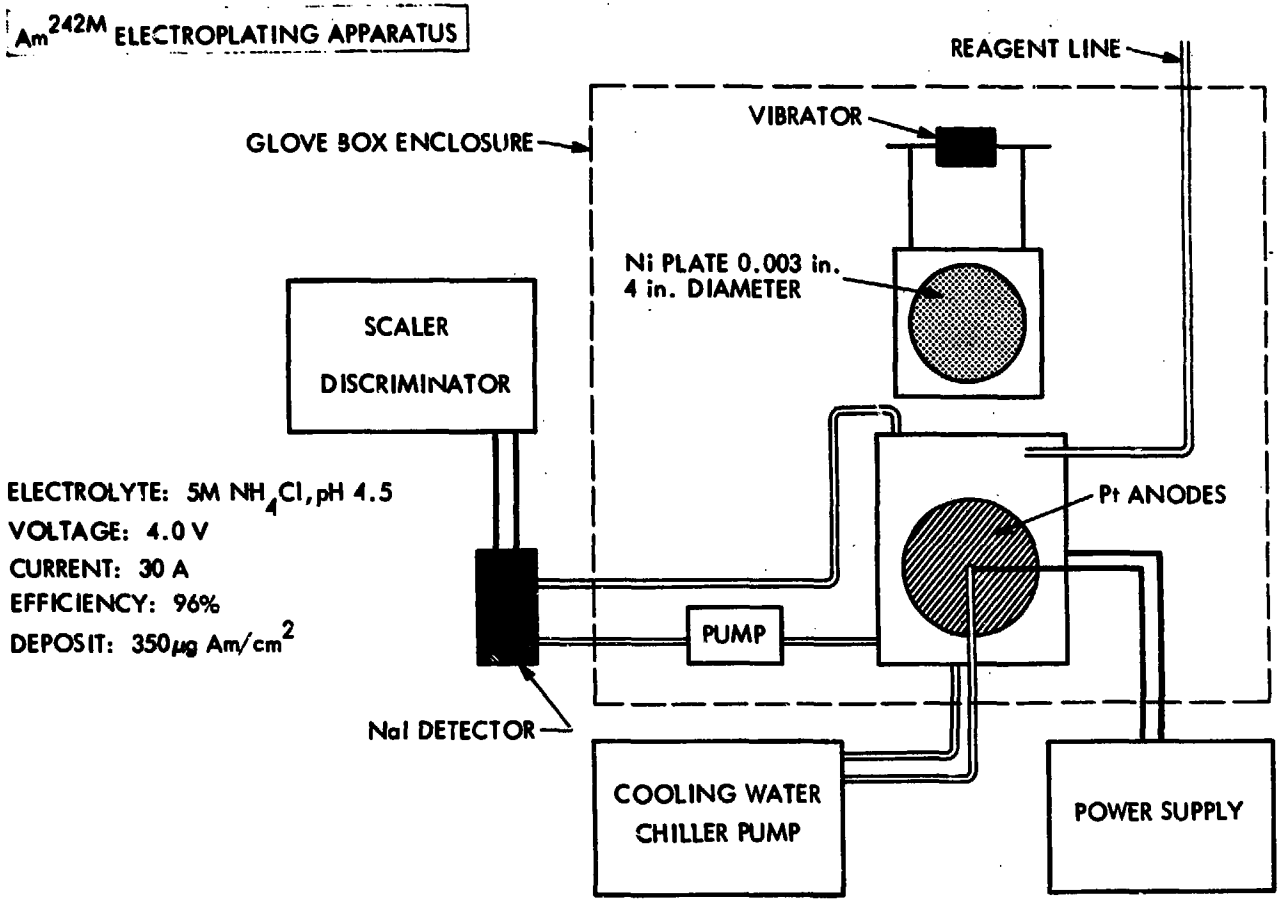

Am^{242M} ELECTROPLATING APPARATUS
REAGENT LINE
GLOVE BOX ENCLOSURE
VIBRATOR
Ni PLATE 0.003 in.
4 in. DIAMETER
SCALER
DISCRIMINATOR
ELECTROLYTE: 5M NH_4Cl, pH 4.5
VOLTAGE: 4.0 V
CURRENT: 30 A
EFFICIENCY: 96%
DEPOSIT: 350μg Am/cm^2
Pt ANODES
PUMP
NaI DETECTOR
COOLING WATER
CHILLER PUMP
POWER SUPPLY

Fig. 1

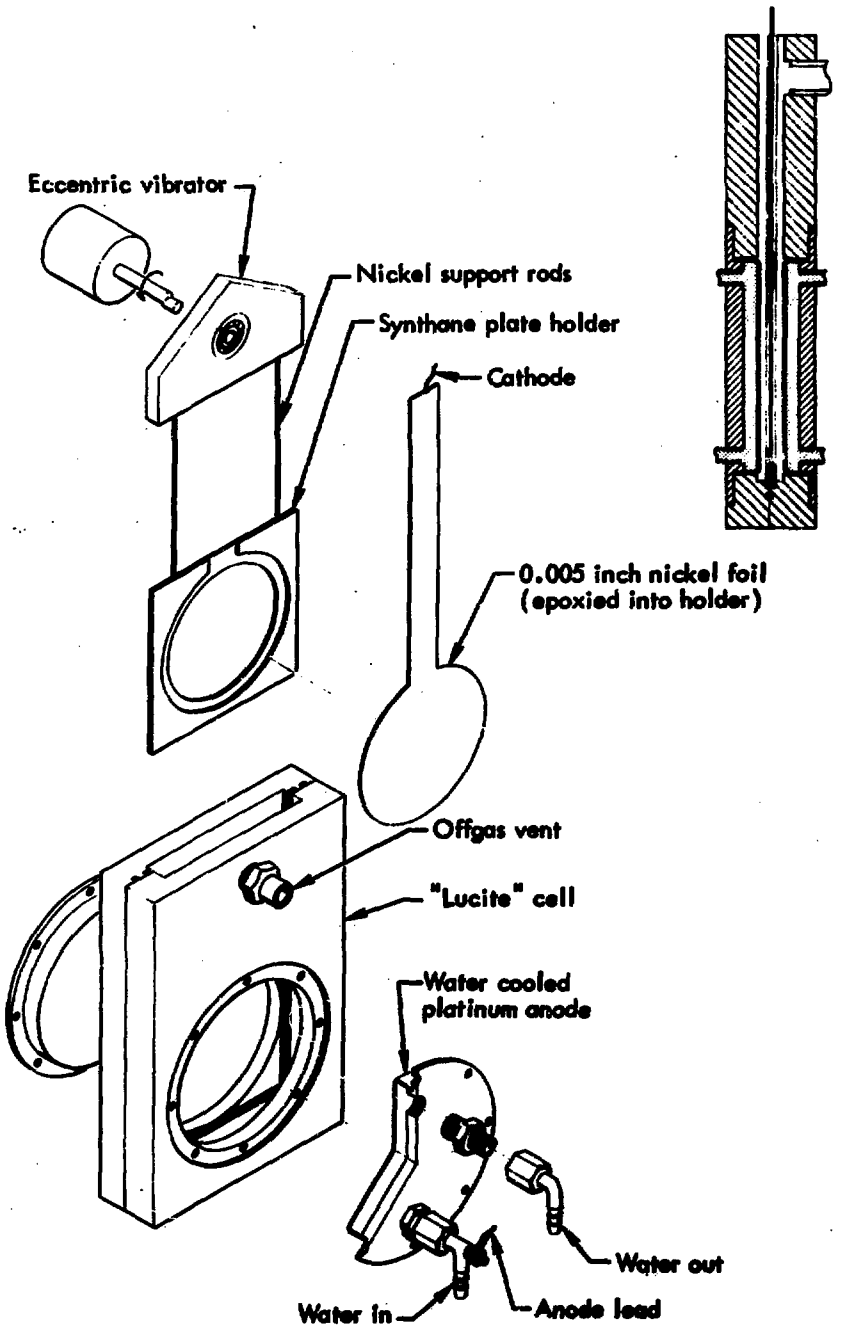

Eccentric vibrator
Nickel support rods
Synthane plate holder
Cathode
0.005 inch nickel foil
(epoxied into holder)
Offgas vent
"Lucite" cell
Water cooled
platinum anode
Water out
Water in
Anode lead

Fig. 2

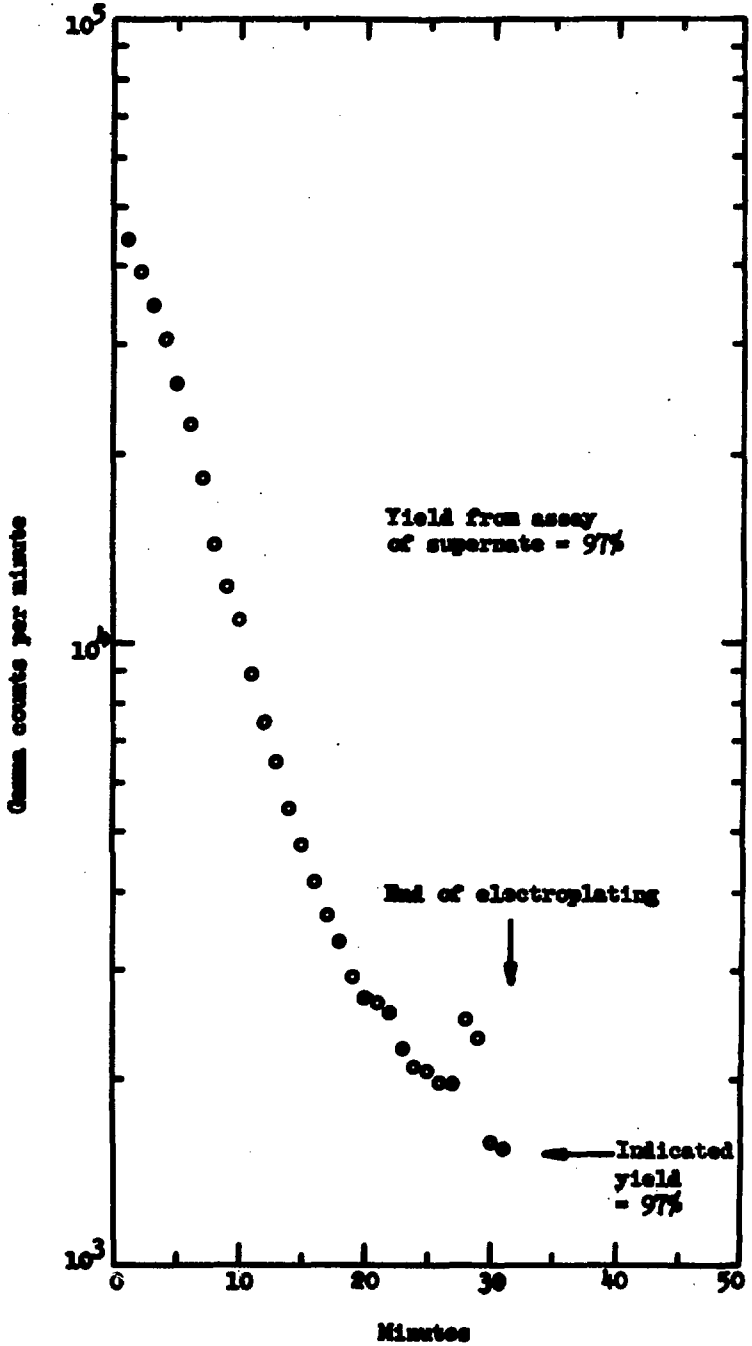

10^5
10^4
10^3
Gamma counts per minute
Yield from assay of supernate = 97%
End of electroplating
Indicated yield = 97%
0
10
20
30
40
50
Minutes

Fig. 3

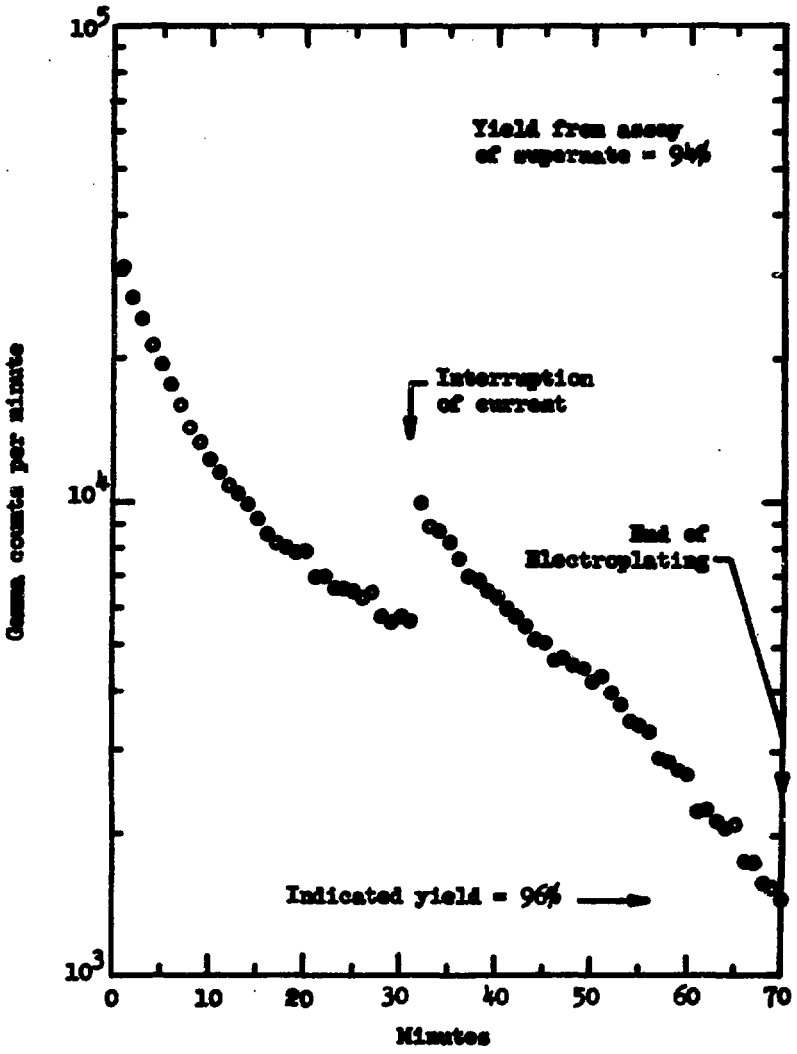

Yield from assay of supernate = 9%
Interruption of current
End of Electroplating
Indicated yield = 96%
Gamma counts per minute
Minutes
10^5
10^4
10^3
0
10
20
30
40
50
60
70

Fig. 4

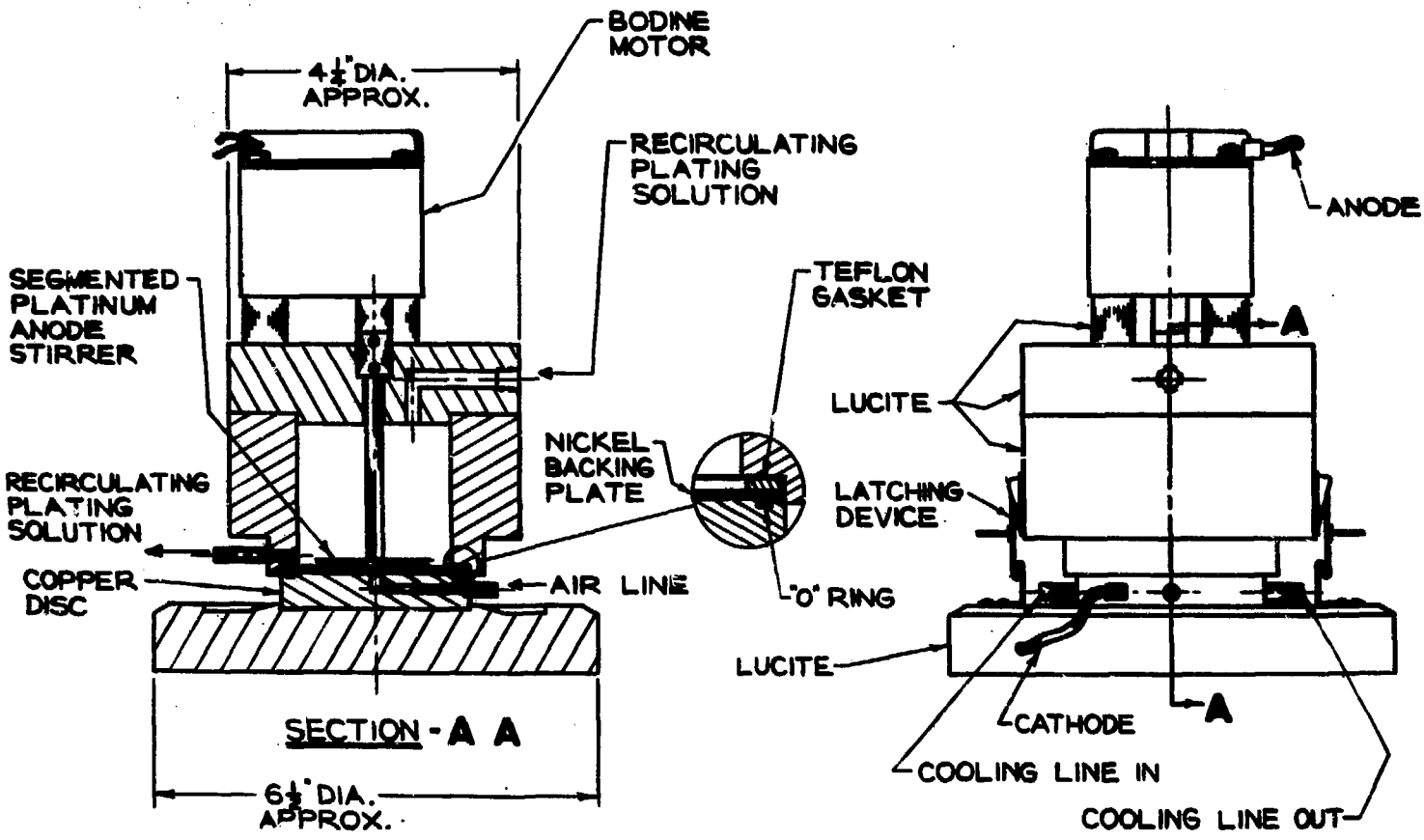

BODINE MOTOR
4½" DIA. APPROX.
RECIRCULATING PLATING SOLUTION
SEGMENTED PLATINUM ANODE STIRRER
TEFLON GASKET
ANODE
A
LUCITE
NICKEL BACKING PLATE
LATCHING DEVICE
RECIRCULATING PLATING SOLUTION
COPPER DISC
AIR LINE
"O" RING
LUCITE
SECTION - A A
CATHODE
A
COOLING LINE IN
COOLING LINE OUT
6½" DIA. APPROX.

Fig. 5

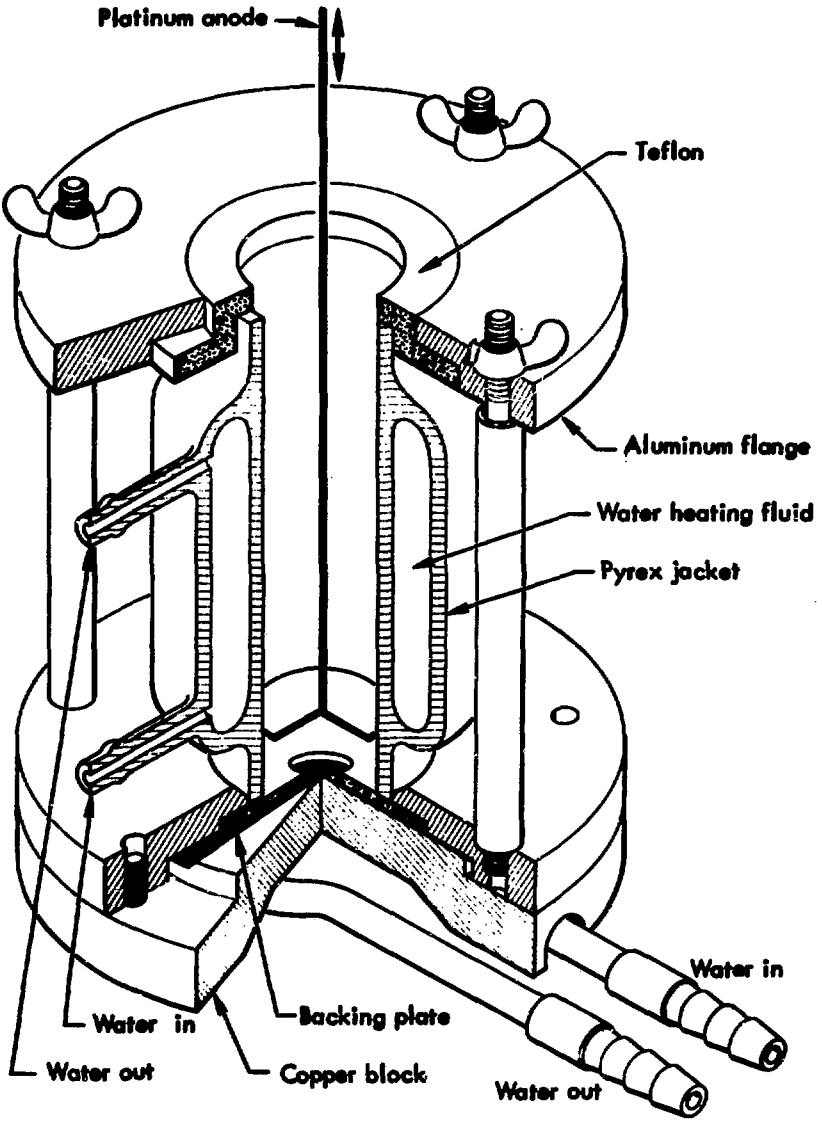

Platinum anode
Teflon
Aluminum flange
Water heating fluid
Pyrex jacket
Water in
Backing plate
Water out
Copper block
Water in
Water out

Fig. 6

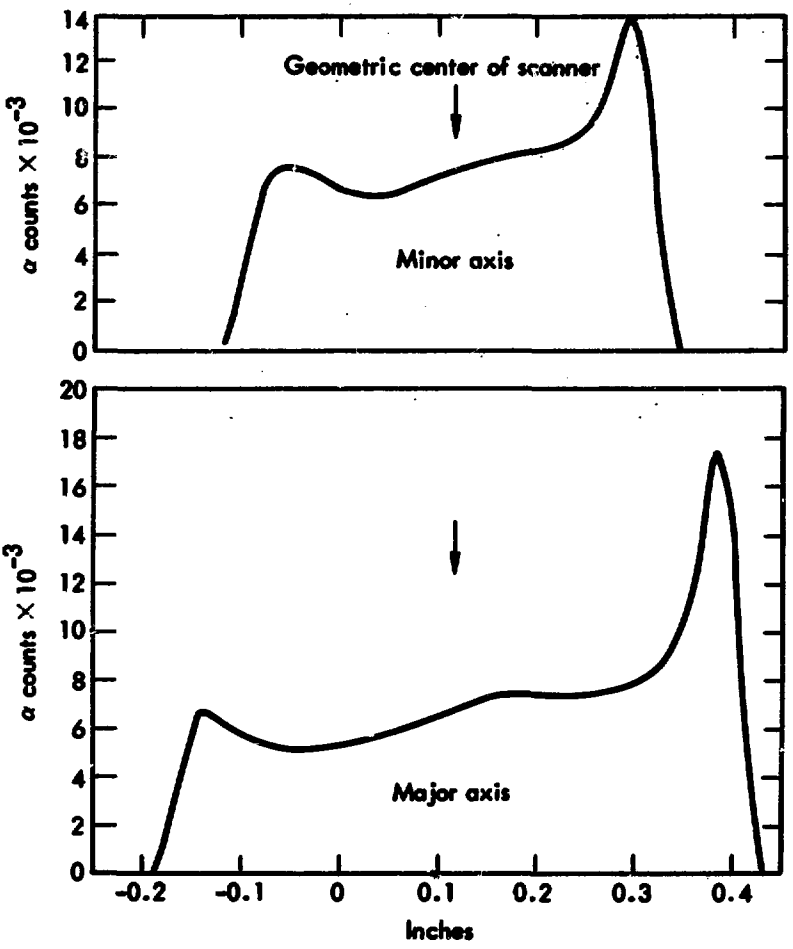
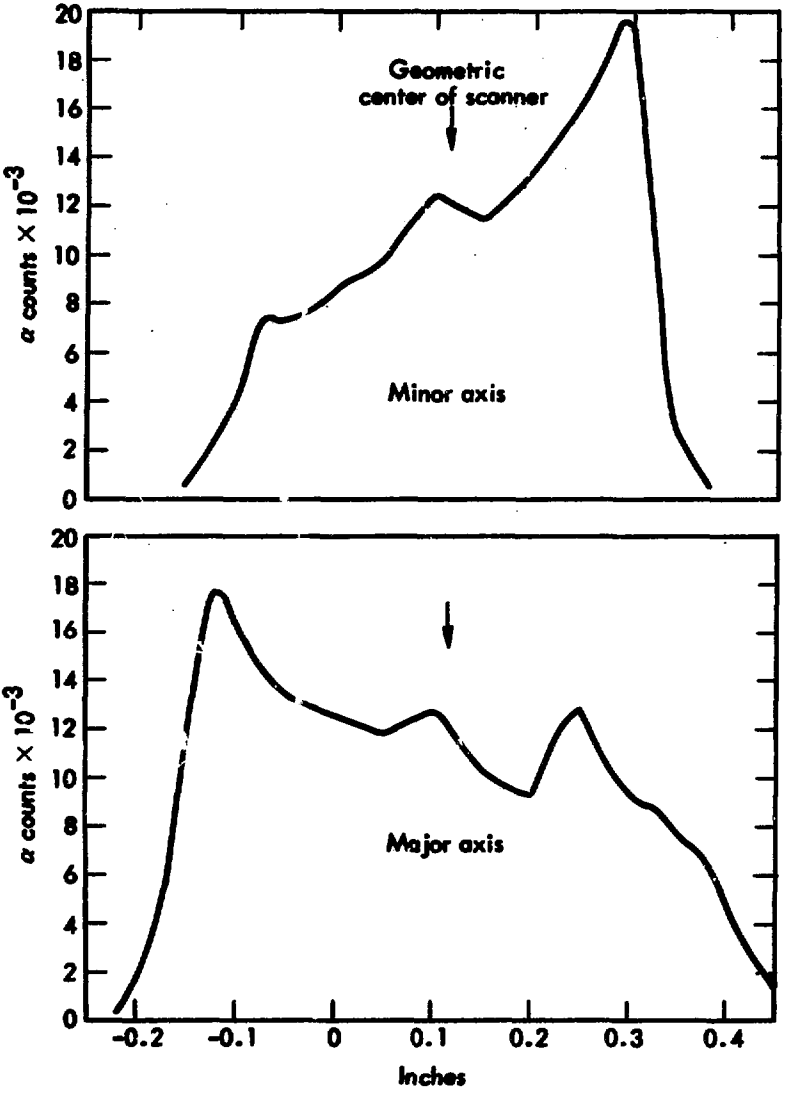

Geometric center of scanner
Minor axis
Major axis
α counts × 10⁻³
α counts × 10⁻³
Inches
0
2
4
6
8
10
12
14
16
18
20
-0.2
-0.1
0
0.1
0.2
0.3
0.4

Fig. 8


Geometric
center of scanner
Minor axis
Major axis
α counts × 10⁻³
20
18
16
14
12
10
8
6
4
2
0
-0.2
-0.1
0
0.1
0.2
0.3
0.4
Inches

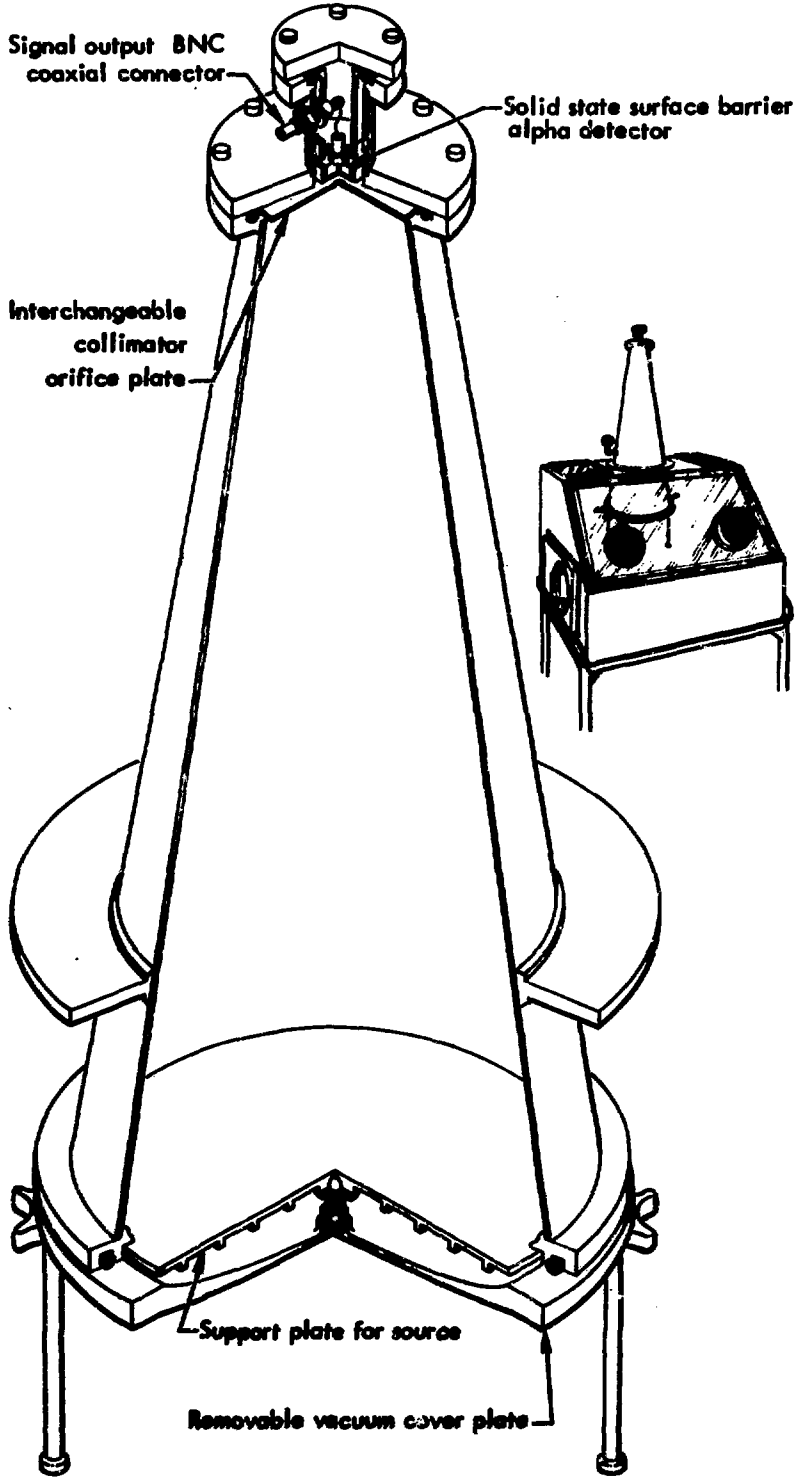

Signal output BNC coaxial connector
Solid state surface barrier alpha detector
Interchangeable collimator orifice plate
Support plate for source
Removable vacuum cover plate

Fig. 10